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THE USE OF THE GAMMA SPECTROMETER FOR THE IDENTIFICATION OF RADIONUCLIDES IN WATER

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THE USE OF THE GAMMA SPECTROMETER FOR THE IDENTIFICATION OF RADIONUCLIDES IN WATER

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The availability of commercial gamma spectrometers has greatly simplified the problem of identification of radionuclides. Experience with this equipment for such identification in rain, cistern, and surface waters is described. Data are presented showing differences obtained as a function of water fraction, age of samples, etc. Information on the applicability of this technique to quantitative determinations of specific radionuclides is also included.

INTRODUCTION

It has been becoming increasingly apparent in past years that the determination of "gross radioactivity" does not adequately characterize the radiological content of water. Identification of the radioactive materials is necessary for a meaningful assessment of the potential health hazard due to the presence of this type of contamination. Gamma scintillation spectrometry offers a possible means of simplifying this identification. In addition, since gamma radiation is affected by self-absorption to only a very minor degree, this technique permits the use of large samples with much less stringent sample preparation than is required for beta or alpha counting.

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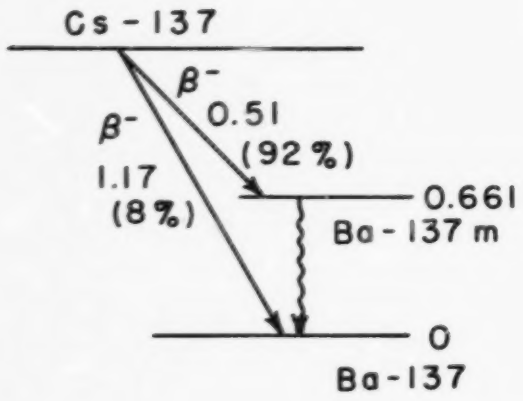
Unfortunately, gamma spectometry is not a panacea for all the problems involved in radioactivity measurements. A number of our more important radionuclides, including strontium-90, strontium-89, yttrium-91, phosphorus-32, and hydrogen-3, either do not emit gamma radiation, or do so in only a small percentage of the radioactive disintegrations. Furthermore, gamma measurements appear to be somewhat less sensitive than alpha or beta measurements, and some interference of one gamma radiation with another occurs in complex mixtures.

PRINCIPLES

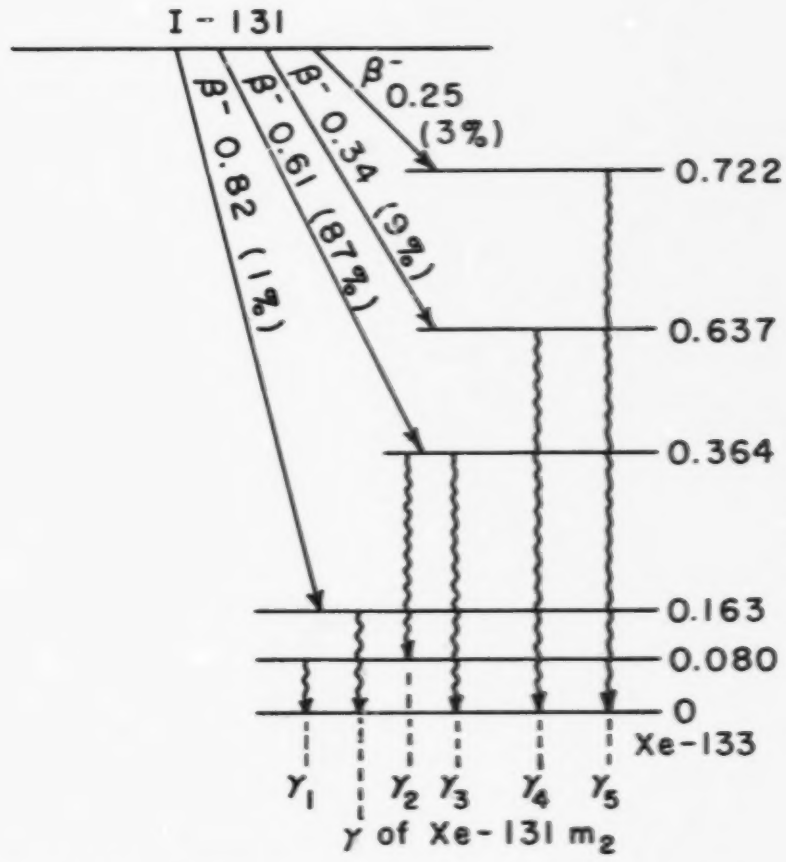
The basis for gamma spectrometry is the emission of gamma photons in a characteristic line spectrum consisting of a few discrete energies. Figure 1 shows the energy levels of cesium-137 and iodine-131, and the gamma radiations emitted. Utilization of these spectra for identification requires detectors capable of producing an output proportional to the gamma energy.

Gamma photons interact with matter in one of three ways, two of which are of importance in gamma spectoscopy: they may produce a secondary (photo-) electron carrying the full gamma energy; they may produce a secondary (Compton) electron carrying less than the full gamma energy plus a secondary gamma photon; or, less important, they may produce a positron-electron pair. Figure 2 illustrates these interactions. The secondary electrons then transfer their energy to the scintillating crystal, producing light photons proportional in number to the energy. By the use of suitable light-collecting and electronic apparatus, a pulse is obtained proportional to the amount of energy lost by the incident gamma photon to the crystal.

Cs - 137

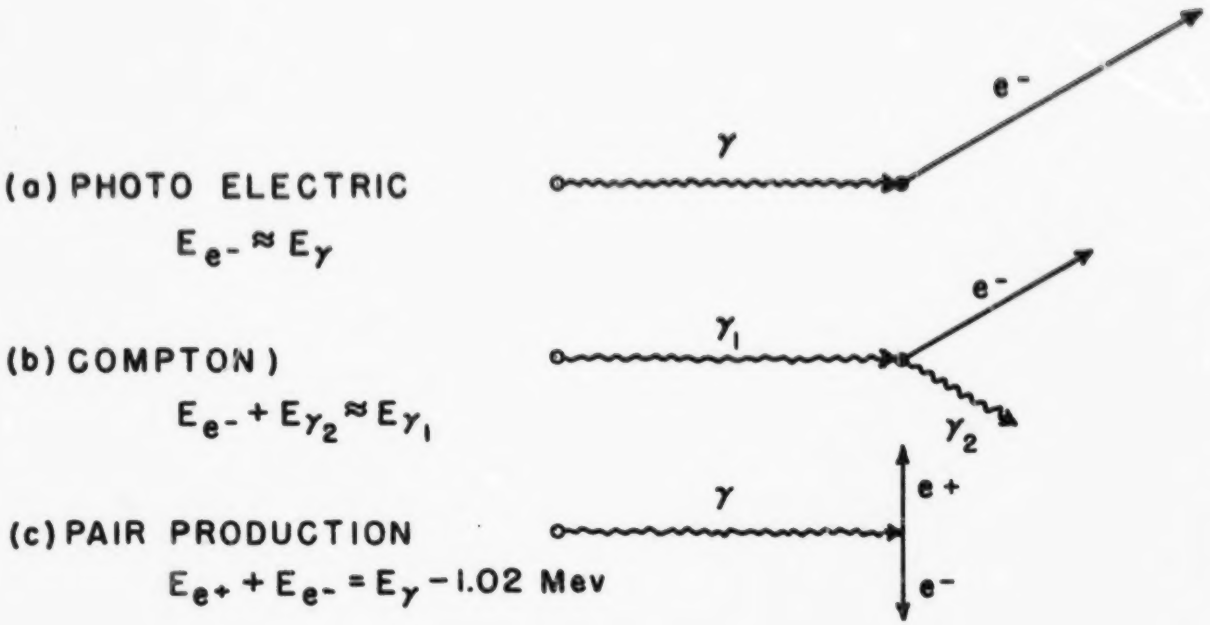


I - 131



ENERGY LEVELS

FIGURE 1



INTERACTIONS OF GAMMA PHOTONS
FIGURE 2

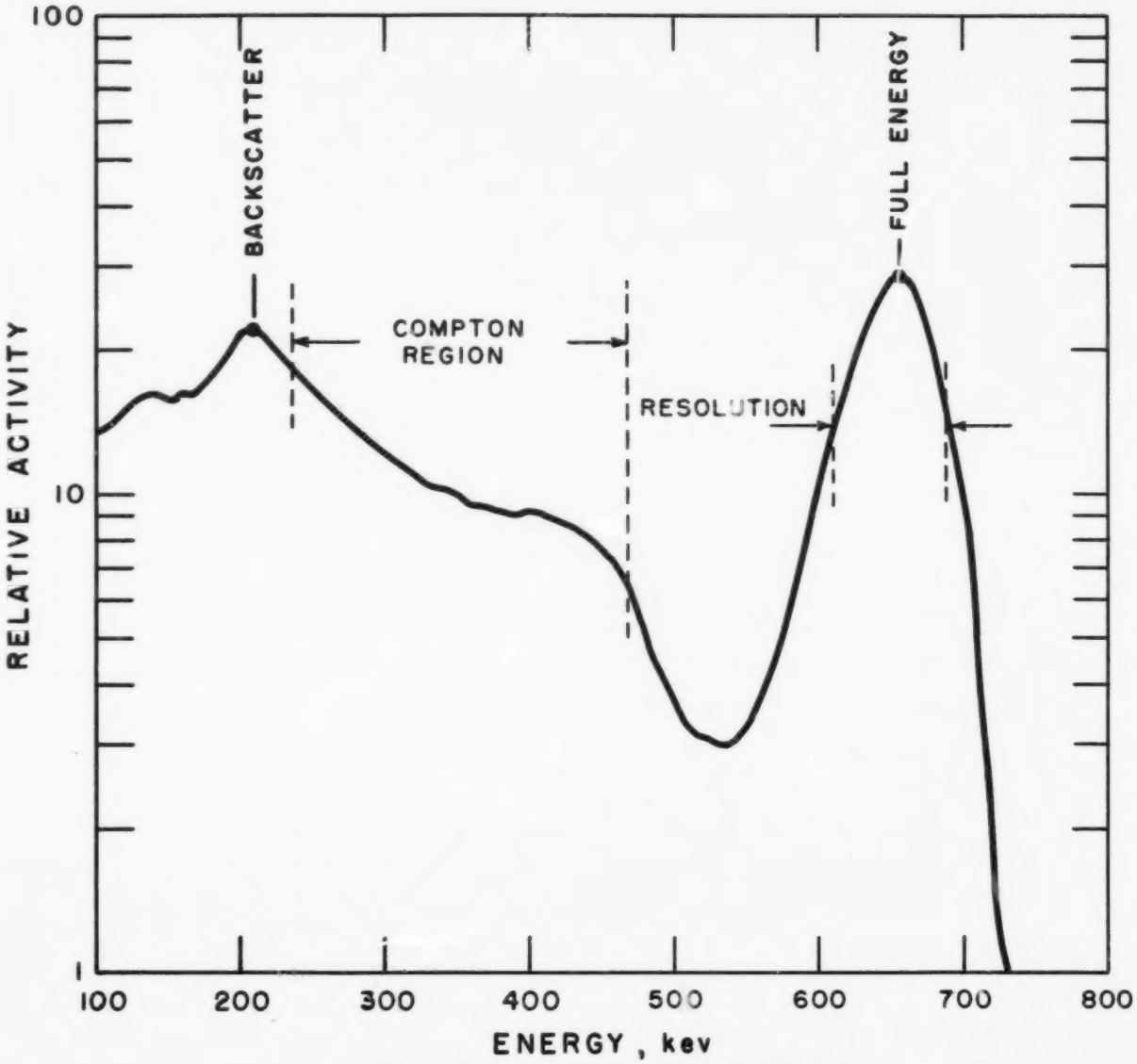
The initial line spectrum of the gamma emitter is thus modified as shown in Figure 3 for the cesium-137 spectrum. There are two factors involved in this spectrum modification--a broadening of the line itself (resolution) and a degradation of the spectrum caused by incomplete capture of the photon energy.

INSTRUMENTATION

Almost all gamma spectroscopy systems now use a thallium-activated sodium iodide crystal, a photomultiplier tube, and a linear amplifier to produce a pulse proportional to the gamma energy. The resulting pulses are then usually fed into a unit which (a) responds to all pulses larger than a preset value (discriminator), (b) responds to all pulses between two preset values (single-channel analyzer), or (c) sorts pulses into a number of channels depending on their size (multi-channel analyzer). Figure 4 illustrates a single-, a 20-, and a 100-channel analyzer. The full spectrum can be obtained with each of these instruments: the multi-channel analyzer gives the full spectrum in a single run or a few runs; the single-channel analyzer requires a channel-by-channel plot; the discriminator requires plotting of the difference between consecutive counts.

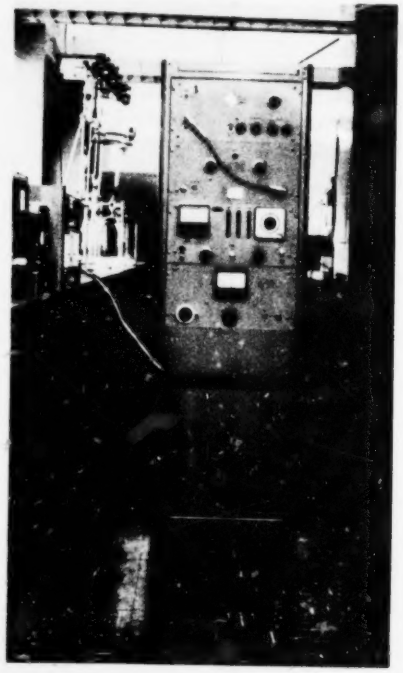
APPLICATION TO WATER SAMPLES

Isotopes: In the samples to be presented here, primary interest was in fission product contamination. Some of the major fission products are listed below, together with their half-lives and characteristic gamma radiations. The more important radiations are listed in bold-face type.

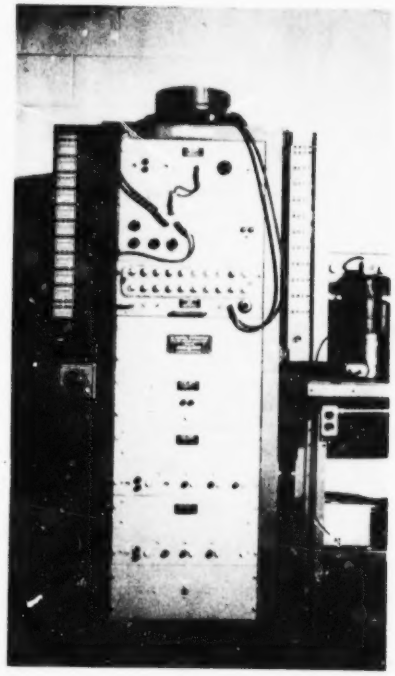


CESIUM-137 SPECTRUM

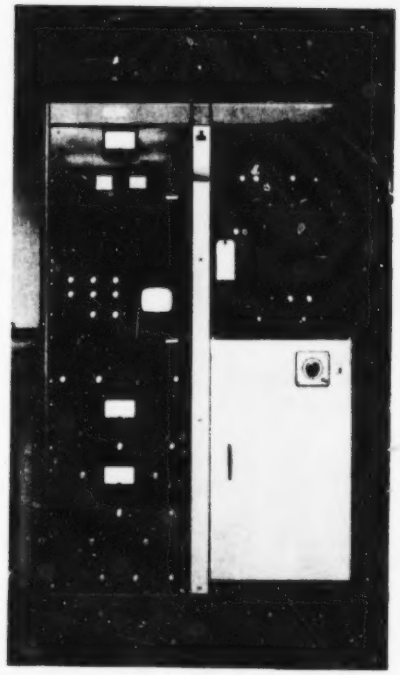
FIGURE 3



SINGLE CHANNEL



20 CHANNEL



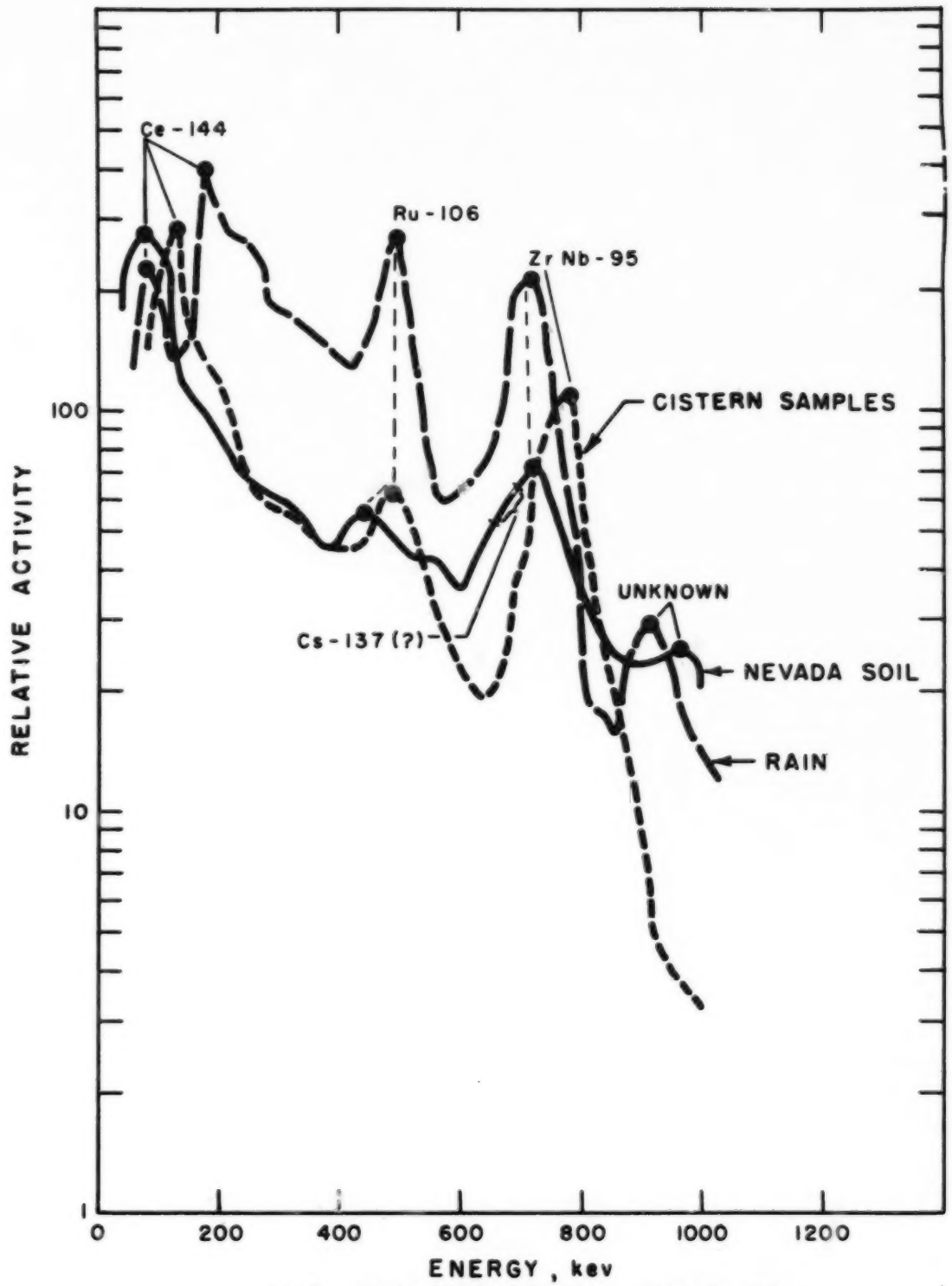
100 CHANNEL

GAMMA SPECTROMETERS

Nuclide	Half-Life	Gamma Radiations, Mev
Strontium-89	53 d	None
Strontium-Yttrium-90	28 y	None
Yttrium-91	58 d	None (<0.5%)
Zirconium-95	63 d	0.72
Ruthenium-Rhodium-103	41 d	0.040, 0.50, 0.61
Ruthenium-Rhodium-106	1 y	0.51, 0.62
Iodine-131	8 d	0.080, 0.28, 0.36, 0.64, 0.72
Cesium-Barium-137	33 y	0.66
Barium-Lanthanum-140	13 d	0.49, 0.82, 0.03, 0.16, 0.30, 0.54, 1.60
Cerium-141	33 d	0.14
Cerium-Praseodymium-144	275d	0.13
Neodymium-147	11 d	0.09, 0.31, 0.39, 0.52

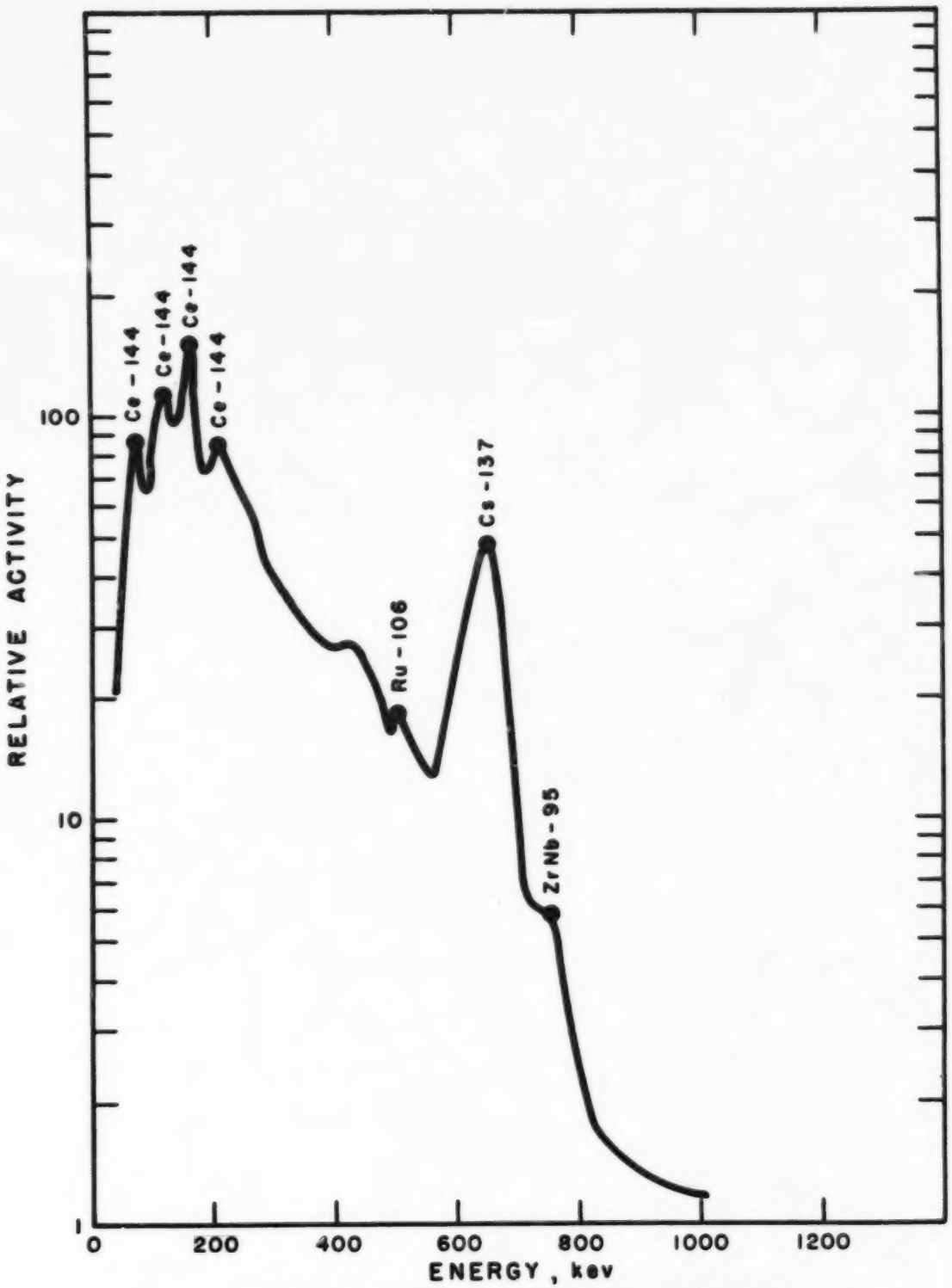
Spectra: Figures 5 through 9 show the gamma spectra obtained on some environmental samples which have been analyzed at the Center. Figure 5 shows spectra for three samples, soil from the Nevada Weapons Testing Area, rain, and cistern water, all of them rather old and with relatively simple spectra. All show a number of isotopes characteristic of fission products of this age--cerium-144, zirconium-niobium-95, and ruthenium-106. In addition, two of the samples show indications of a peak at about 950 kev energy. This radiation, which is not characteristic of any of the fission products, has not been identified by the Center. It may very well be due to induced radioactivity.*

* Scientists of the U. S. Naval Radiological Defense Laboratory have reported finding cobalt-60 and manganese-54, induced radioactivities, in fallout materials.



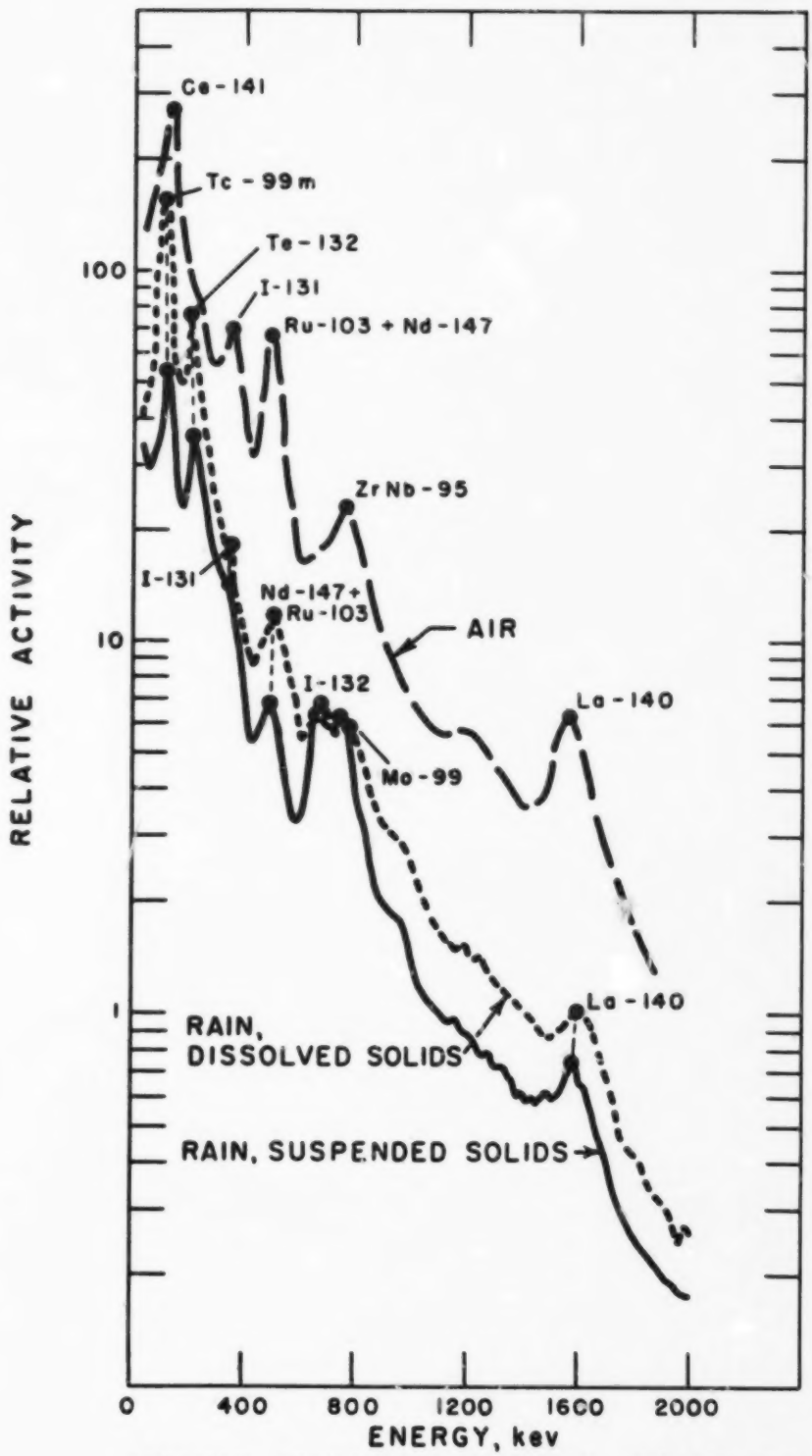
OLD ENVIRONMENTAL SAMPLES

FIGURE 5



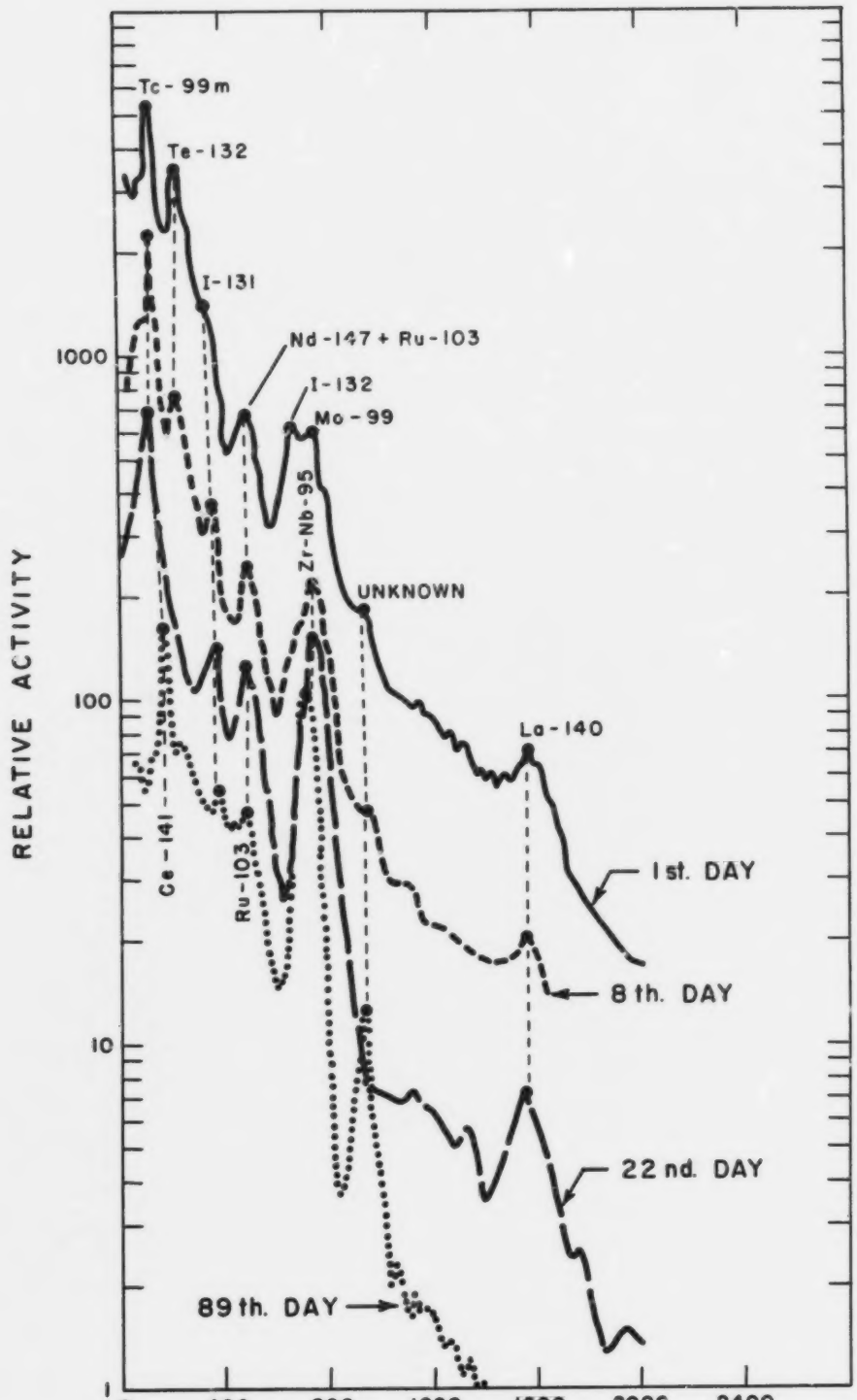
OLD CISTERN SLUDGE SAMPLE

FIGURE 6



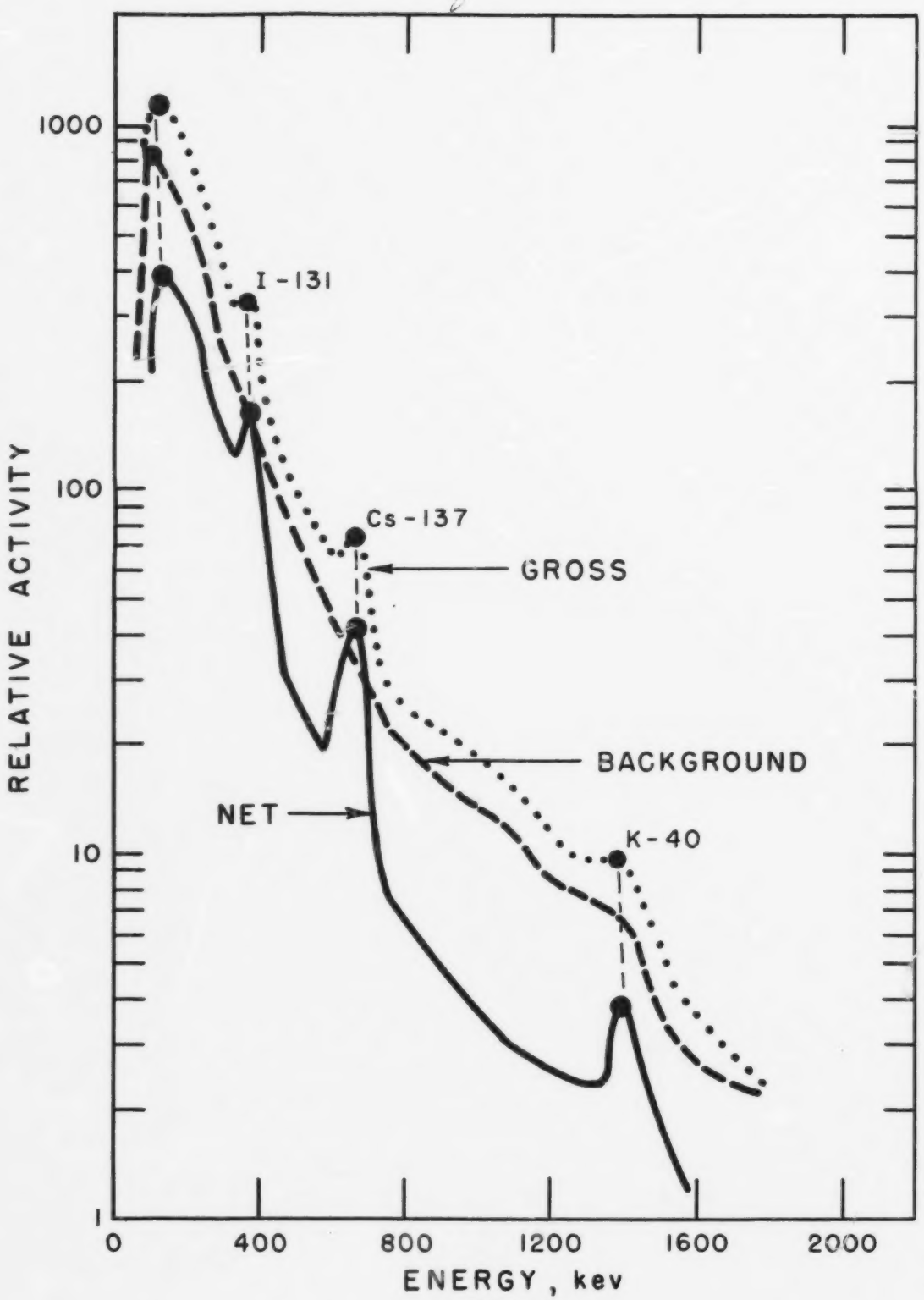
YOUNG ENVIRONMENTAL SAMPLES

FIGURE 7



RAIN SAMPLE DECAY

FIGURE 8



MILK

FIGURE 9

Figure 6 illustrates the spectrum obtained from a sample of cistern sludge approximately 18 months after collection. Because of the concentration of isotopes in this material compared to those in water samples, the gamma peaks are more distinct. The predominant nuclides are cerium-144, and cesium-137, with a small peak in the region corresponding to ruthenium-106, and a step in the curve which is probably indicative of a trace of zirconium-niobium-95. These findings are in reasonable agreement with the predicted abundances (1) which at this age show approximately 65% of total activity due to cerium-144, some 6% to cesium-137, about 5% to ruthenium-106, and about 4% to zirconium-niobium-95. The remaining activity is chiefly from promethium-147 and strontium-90 which do not emit gamma radiation. This spectrum does not show any peaks due to induced activity.

Figure 7 shows the spectra of three relatively young environmental samples--one, particulate matter in air, and two, suspended and dissolved fractions of rainfall.

The air sample shows peaks due to cerium-141, iodine-131, ruthenium-103, zirconium-niobium-95, and lanthanum-140. The ruthenium-103 peak at about 500 kev probably includes some neodymium-147, 520 kev. The spectra of the rain samples, which qualitatively are very much alike, show a number of peaks characteristic of fission products of more recent origin. This observation is in accord with the gross beta decay of this material, which indicates that it consists chiefly of fission products less than one week old. Two of the readily identifiable fission products of short half-life are molybdenum-99, with gamma energies of 740 kev and 180 kev, and its daughter, technetium-99m with a gamma of 140 kev. Another parent-daughter pair of short half-life is tellurium-132, with a peak at 220 kev, and

iodine-132, with a gamma energy of 690 kev. Other peaks can be attributed to neodymium-147 plus ruthenium-103, to iodine-131, and to lanthanum-140. The cerium-141 activity is obscured by the low-energy molybdenum and technetium radiations, and the zirconium-niobium activity is masked by the iodine-132 and molybdenum-99 peaks.

Figure 8 illustrates a series of measurements made over a period of time on one of the rain samples included in Figure 7. These curves show the disappearance of the short-lived isotopes molybdenum-technetium-99 (m) and tellurium-iodine-132. Other isotopes of intermediate half-life, typified by iodine-131 and lanthanum-140, increase in prominence and then die out. Finally, isotopes of longer half-life become the most prominent. Among these are zirconium-niobium-95 and the unknown radionuclide at 950 kev which was noted in the curves of Figure 5. Two peaks, while remaining prominent throughout, have changed in nature. These are the low energy peak at about 140 kev, initially due mostly to molybdenum-99 and technetium-99m, and finally to cerium-141, and the peak at about 500 kev, which changed from predominantly neodymium-147 to ruthenium-103. Where such mixtures occur, chemical separations are needed for determination of their relative quantities, although sometimes sufficient information can be obtained from decay measurements.

Figure 9 gives a spectrum obtained from milk and also shows the spectrum of instrument background. It is apparent that the activity in the milk represents only a very small increase over the background activity; however, by subtracting the background, a net spectrum is obtained which rather clearly shows three peaks. Most definite are the peaks due to

cesium-137 and iodine-131; there is a reasonably certain identification of a peak due to the natural potassium content of the milk, although the crystal used in these measurements is too small to detect this high-energy (1.46 Mev) radiation with good efficienty. The peak at low energy is probably not due to any radioisotope; it is more likely a combination of the poor statistics caused by the high background in this region and of pileup of the Compton and backscatter from the potassium, cesium, and iodine activities. From such a net curve, the quantitative determination of at least the cesium and iodine activities would be possible, provided the instrument was first calibrated with the individual isotopes and finally with mixtures of the two.

DISCUSSION AND SUMMARY

The principle on which gamma scintillation spectoscopy is based is the emission of a characteristic line spectrum of gamma radiation by a specific radionuclide. By the use of suitable instrumentation, one may obtain an actual spectrum with a peak at the characteristic gamma energy.

This paper has shown the application of gamma spectroscopy as a qualitative tool in the identification of radionuclides in water and in some other environmental samples, especially as applied to fission products. Decay measurements are often useful in confirming tentative identifications. Where further information is needed, quantitative data may be obtained, especially in relatively simple cases, if the equipment has been previously calibrated with radionuclides of known concentration. The qualitative data may be used as a guide for radiochemical separation or alternatively preliminary chemical separations may be used to obtain simplified spectra for quantification.

Although gamma detectors are somewhat less sensitive than beta detectors, this disadvantage is nullified to a great extent by the practicability of using very large initial samples.

ACKNOWLEDGEMENT

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